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Optical and holographic characteristics of photopolymer layers

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In the present work the optical and holographic characteristics of acrylamide-based photopolymer layers are studied. For the first time the refractive index change of a liquid acrylamide photopolymer due to exposure at 532 nm is obtained using a critical angle laser micro-refractometer. The 30 μm thick solid photopolymer films are prepared by casting on glass substrates. Bragg holographic gratings with spatial frequencies of 710 mm^{-1} , 1050 mm^{-1} and 1600 mm^{-1} are recorded using a diode laser operating at 532 nm wavelength. The diffraction efficiency dependence on the exposure energy is investigated. The obtained results are compared with the Stetson holographic recording method, where two gratings are simultaneously recorded in the same location with spatial frequencies 2020 mm^{-1} and 3670 mm^{-1} , using a totally reflected reference wave from the air-photopolymer interface. Despite the fact that in the second method the two gratings share the same dynamic range, higher diffraction efficiencies are observed.

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Keywords: Photopolymers, Refractive index, Holography

1. Introduction

Photopolymers have found application in holographic sensors [1, 2], holographic data storage [3, 5], fabrication of holographic optical elements [6], security holograms [7], electro-optical switchable devices [8], and also some non-imaging applications such as adhesives, coatings and inks [9]. Because the lasers and the optical systems for work in the UV are very expensive, the majority of the photopolymers have been designed to respond to visible light. The basic formulation of the dry photopolymer system contains photoinitiator and monomer that are dispersed in a binder matrix. In the investigated acrylamide-based photopolymer system photoinitiation is a two step process. Upon illumination of the photopolymer with light of appropriate wavelength the sensitizing dye absorbs a photon and reacts with an electron donor to produce free radicals (initiation step). These initiate polymerization where the light was absorbed. Chain propagation or termination follows the initiation. Currently accepted models describe the recorded pattern formation as a result of changes in the density and

the molecular polarizability, which accompany the polymerization. The models [10-12] predict that the key factor controlling the dynamics and final properties of the recorded hologram (refractive index space profile and modulation) is proportional to the ratio of polymerization rate and monomer diffusion rate. Both parameters are strongly dependent on the nature of the photopolymer system and having separate information about the diffusion and polymerization rates is important for the optimization of the photopolymer system. We have previously characterized the monomer and short polymer chains diffusion rates at the initial stage of holographic recording in acrylamide base-photopolymers [13] and also the polymerization rate during homogeneous illumination utilising Raman spectroscopy [14]. In the present investigation we have focused our attention on the characterization of the polymerization rate by determination of the photoinduced changes of the surface refractive index during homogeneous illumination and polymerization. The surface refractive index is measured at 532 nm wavelength by a laser refractometer. The properties of the holographic recording using two different geometries are also characterised and compared.

2. Experimental details

2.1. Sample preparation

A stock unsensitised photopolymer solution was prepared by mixing 2.4 g of acrylamide, 0.75 g of N,N'-methylene bisacrylamide, 4.5 ml of triethanolamine and 52.5 ml of (10 wt.% PVA stock solution). Erythrosine B stock solution of 0.11 % w/v concentration was prepared by adding 0.11 g of dye into 100 ml distilled water. In order to prepare sensitised layers, 5ml of photopolymer stock solution were mixed with 0.7 ml and 1 ml of dye stock solution resulting in dye concentrations of 0.7 wt.% and 1 wt.% respectively. For surface refractive index measurements liquid layers were prepared by depositing 0.3 ml on a glass slide. The solid layers were obtained by casting 0.3 ml on microscopic slides and dried for 24 hours in dark.

2.2. Optical measurements

The transmission spectra of the solid samples were measured with 0.01 % experimental uncertainty in the 400 nm - 800 nm spectral region with a Cary 5E spectrophotometer. The refractive index (RI) was measured with the laser micro-refractometer at 532 nm, i.e. in the absorption band. The method was earlier described in details in [15, 16]. Briefly, the determination of the critical angle is carried out with the help of a metal grating allowing observation if total internal reflection at the prism/photopolymer layer interface occurs. The photopolymer layer is located between a high refractive index ($n_p > 1.5$) prism and the grating. At lower incidence angles the laser beam is transmitted through the sample and the grating, and a diffraction pattern is observed. At critical angle incidence, total internal reflection occurs, no beam passes through the grating and the diffraction pattern disappears. The experimental uncertainty mainly depends on the accuracy of the goniometer used - a "Microcontrol" rotary stage with 1 arcmin resolution. With this experimental refractometer the RI was measured with ± 0.0002 experimental uncertainty for liquid samples, and ± 0.0005 for solid samples, respectively.

2.3. Holographic recording set-up

As illustrated in Fig.1, a symmetrical two-beam interference set-up was used for holographic grating recording in Bragg regime with three different angles 2θ - 21.7° , 32.24° and 50.33° between the recording beams of light. The

corresponding spatial frequencies were 710 mm^{-1} , 1050 mm^{-1} , and 1600 mm^{-1} . A low-intensity He-Ne laser was employed as a read-out beam to monitor the buildup dynamics of the grating during the holographic recording.

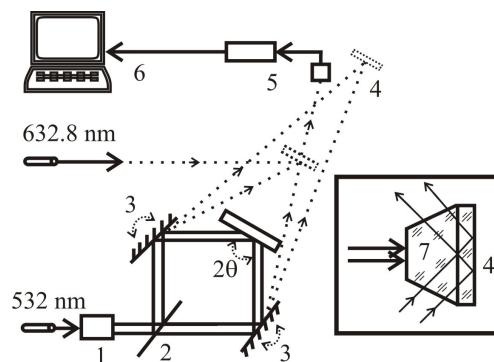


Fig. 1. Holographic setup: 1- beam expander; 2- beam splitter; 3- mirrors; 4- photopolymer; 5- powermeter; 6- computer; 7- glass prism

The total laser beams' intensity was 10 mW/cm^2 . Two high spatial frequency gratings were simultaneously recorded in Stetson regime, using the holographic setup [17], presented in the inset of Fig.1. The reference beam was totally reflected from the photopolymer - air interface at the 65° incidence angle. The object beam was incident normally on the photopolymer. Transmission and reflection gratings were simultaneously recorded with spatial frequencies 2020 mm^{-1} and 3670 mm^{-1} , respectively.

3. Results and discussion

The transmission spectra for 0.7 wt% and 1.0 wt% dye concentration are presented in Fig.2.

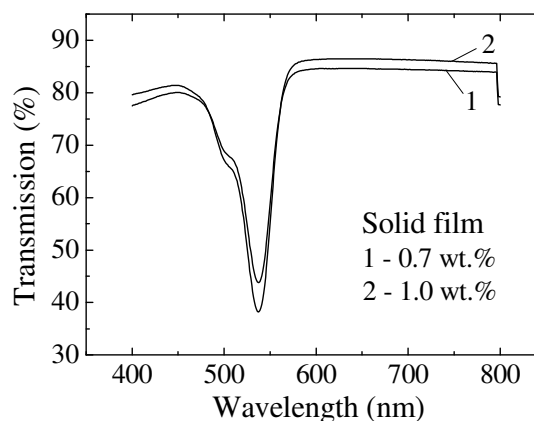


Fig. 2. Transmission spectra dependence on the dye concentration

The photopolymer is optimised for recording at 532 nm and the sensitizer (Erythrosine B) has maximum absorption at 535 nm.

The results of RI measurements of liquid photopolymer layers characterized by different dye concentrations after delivery of exposure at 532 nm are presented in Fig. 3. It is seen that, as could be expected, the RI increases after illumination.

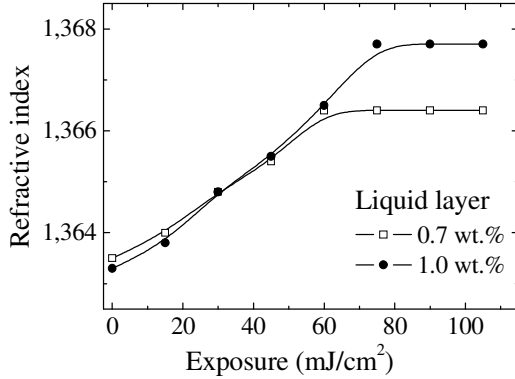


Fig. 3. Evolution of the refractive index of liquid layers versus exposure

Starting from $RI = 1.3632 \pm 0.0002$, for the both cases the RI increases without delay and achieves different saturated values at different exposure levels, depending on the dye concentration. Despite the 0.3 wt% difference in dye concentration, the saturated RI values for 1 wt% are about 0.001 higher revealing that this dye concentration provides more efficient polymerization and larger RI change.

Quite different is the RI dependence on exposure for the solid films, illustrated in Fig. 4.

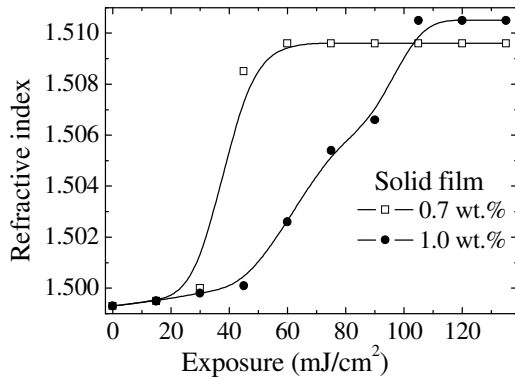


Fig. 4. Evolution of the solid film's refractive index versus exposure

For the dye concentration 0.7 wt% threshold exposure 30 mJ/cm² is observed and 45 mJ/cm² for

1 wt%. The RI saturation exposure 60 mJ/cm² for 0.7 wt% is the same for the film and the liquid layer. The results of the RI investigations are summarized in Table 1.

Refractive index difference is:

$$\Delta RI = RI_{E=0} - RI_{E=E_{saturation}} \quad (1)$$

Table 1. Summarized results from the refractive index measurements

Sample	C, wt. %	$E_{saturation}$ mJ/cm ²	Saturated RI, $\pm 2.10^{-4}$ ($\pm 5.10^{-4}$)*	ΔRI
Liquid layer	0.7	60	1.3664	0.003
	1	75	1.3677	0.004
Solid film	0.7	60	1.5096*	0.010
	1	105	1.5105*	0.011

The observed differences in the RI dependence on the exposure and dye concentration for liquid formulations and solid films can be explained having in mind that penetration depth in the total internal reflection is about 1 μm [18].

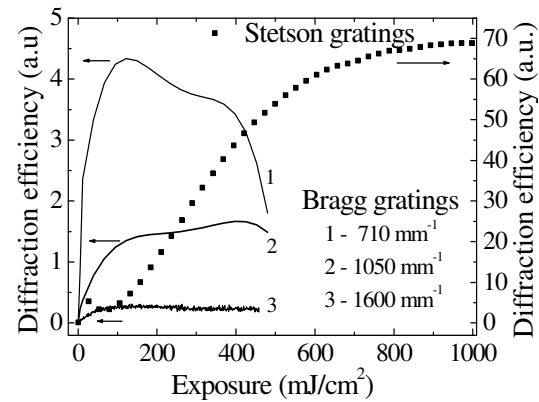


Fig. 5. Diffraction efficiency dependence on exposure

Near the boundary between glass prism and liquid phase the surface tension is smaller, compared with the surface tension at the glass substrate - film boundary. In this case the RI change

due to photopolymerization is observable immediately, in less than 0.1 sec. On the other hand, near the solid film surface, this process is delayed due to the higher surface tension. The difference between saturated RI values of solid film and liquid layers for the two different concentrations is similar - 0.143 ± 0.001 and in this concentration range does not depend on the dye concentration.

The diffraction efficiency (DE) dependence on the exposure is shown in Fig. 5. It is interesting to note, that DE for the transmission Bragg gratings is much lower compared with two gratings DE, simultaneously recorded by Stetson's method in the same location.

In order to explain this effect, two important facts should be taken into account: 1. The two Stetson gratings are of different type (reflection and transmission) and their angles of reconstruction and diffraction coincide. 2. The dynamic range of the RI modulation (Δn) is large enough in order to be shared and to achieve high DE.

From Kogelnik's coupled wave theory [19], for 100 % DE one needs a RI modulation, according to the following relation:

$$\Delta n = \frac{\lambda \cos \theta}{2d} \quad (2)$$

For $\theta = 30^\circ$, $\lambda = 532 \text{ nm}$ and $d = 30 \text{ }\mu\text{m}$, the needed RI modulation is $\Delta n = 0.0077$.

From the Table 1 it is seen that the RI change is enough to ensure simultaneous recording of two grating with relatively high DE.

4. Conclusion

In summary, in the present work the refractive index changes under illumination of liquid and solid photopolymer layers at 532 nm is measured with a laser microrefractometer. In solid films this change is approximately twice that in liquid layers. The investigated photopolymer can be used for holographic recording not only as a solid film, but also as liquid recording medium. This could prove useful in the development of photopolymerisable nanocomposites where high permeability is required in order to achieve redistribution of the nanodopants and consequently improvement of the recording dynamic range.

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